SPECIFICATION

TO ALL WHOM IT MAY CONCERN:

          BE IT KNOWN THAT WE, HIIDEAKI YAMASAKI, a citizen
of Japan residing at Nirasaki-Shi, Yamanashi, Japan, YUMIKO
KAWANO, a citizen of Japan residing at Nirasaki-Shi, Yamanashi,
Japan, have invented certain new and useful improvements in

          FILM-FORMATION APPARATUS AND SOURCE SUPPLYING APPARATUS
          THEREFOR, GAS CONCENTRATION MEASURING METHOD

of which the following is a specification:-

- 1 -

TITLE OF THE INVENTION

FILM-FORMATION APPARATUS AND SOURCE
SUPPLYING APPARATUS THEREFOR, GAS CONCENTRATION
MEASURING METHOD

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BACKGROUND OF THE INVENTION

The present invention generally relates to
film-formation apparatuses, and more particularly to
a CVD apparatus that can monitor and control the
10 source gas concentration by way of an infrared
spectrometer.

CVD is an indispensable film-formation
technology in the fabrication process of
semiconductor devices.

15 In the film-formation process according to
CVD (chemical vapor deposition), particularly MOCVD
(metal-organic chemical vapor deposition), in which
formation of film is achieved by using an MO (metal-
organic) source, a liquid source compound that
20 contains the constituent elements of the film to be
formed, or a liquid source prepared by dissolving a
solid source compound containing such constituent
elements into a solvent, is transported to a
vaporizer located near to the processing vessel.
25 There, the vaporizer causes vaporization of the
source compound thus transported and there is
produced a source gas as a result. The source gas
thus produced is then introduced into a processing
vessel of the CVD apparatus, and desired formation of
30 a film such as an insulation film, a metal film or a
semiconductor film, is achieved in the processing
vessel, by causing decomposition of the source gas.

In the MOCVD process, on the other hand,

- 2 -

there are cases in which a liquid source compound or a solid source compound has to be vaporized in a bubbler. In such a case, the source gas formed as a result of the bubbling is transported to the

5 processing vessel via a source gas line. In such a case, there is a need of controlling the concentration of the source gas by controlling the flow rate or pressure of the source gas in the gas line for obtaining a film of desired quality.

10 In the case using a vaporizer, the vaporizer can be provided adjacent to the processing vessel or inside the processing vessel, and the concentration of the source gas supplied to the processing vessel can be controlled by merely

15 controlling the amount of the liquid to be supplied to the vaporizer. There is no particular need of direct detection and monitoring of the concentration of the source gas supplied to the processing vessel.

In the case of supplying a source gas from

20. a bubbler to the processing vessel via a source gas line, too, the concentration of the source gas supplied to the processing vessel has been easily adjusted by merely controlling the carrier gas flow rate or pressure. Thus, there has been no particular

25 need of direct detection and monitoring of the source gas concentration supplied to the processing vessel in the conventional CVD technology.

In the case of forming high-K dielectric films or ferroelectric films, which are used in

30 recent advanced semiconductor devices, or in the case of forming a tungsten (W) film or a ruthenium (Ru) film used also in these semiconductor devices, on the other hand, the vapor pressure of the source gas

- 3 -

obtained from a source material is generally very low, and there can be a case in which the amount of the source gas sufficient for applying an ordinary source gas concentration control, which relies upon control

5   of vaporization of the source gas, cannot be achieved by way of vaporization of the source material.

Thus, in the case of conducting a CVD process with such a low vapor pressure source material, a very small amount vapor produced by

10 holding the source material at a predetermined temperature is supplied to the processing vessel as a source gas by using a carrier gas. Thereby, there occurs extensive dilution of the source gas by the carrier gas, and there arise a case in which accurate

15 determination of the source gas concentration for the source gas actually introduced into the processing vessel is difficult.

Particularly, in the case of conducting a desired CVD process while using a solid source

20 compound of low vapor pressure, there may occur a change of state of the source material with consumption of the source material. Such a change may occur, particularly with regard to the effective surface area of the source material contacting with

25 the carrier gas. When such a change of surface area is caused in the source material, it is generally not possible to avoid significant fluctuation of source gas concentration. Further, such a solid source material has a tendency of forming a temperature

30 distribution inside because of poor heat conduction, contrary to the case of using a liquid source material. This also contributes to the tendency of deviation of the source gas concentration from the

- 4 -

proper concentration range.

Further, in the case of using a liquid source material, too, the fluctuation of the source gas concentration can provide profound effect on the
5   process, as long as the vapor pressure of the source compound is low.

Thus, in such recent technology of MOCVD, direct detection of the source gas concentration is becoming a major issue.
10   As noted above, it is preferable to measure or monitor the source gas concentration directly when conducting a CVD process when such a low-vapor pressure compound is used for the source material. On the other hand, the conventional gas concentration
15   measurement method, such as the one that uses acoustic emission (AE) or specific heat, has a drawback in that reliable measurement is not possible when the measurement is conducted under a low-pressure environment such as the pressure of 50 Torr
20   (6660Pa) or less. Thus, such a conventional measurement process of gas concentration is not applicable to the case of the CVD film-formation process conducted while using a low-vapor pressure source material such as the MOCVD process.
25   Meanwhile, there is proposed a film-formation apparatus disclosed in the Japanese Laid-Open Patent Publication 2001-234348 that is capable of measuring the source gas concentration directly by Fourier-transform infrared (FTIR) spectrometer. This
30   prior art film formation apparatus also controls the gas flow rate based on the result of measurement of the source gas concentration.

In this conventional film-formation

- 5 -

apparatus and method, the mixing ratio of plural gas species is measured by FTIR, and the mixing ratio is adjusted by controlling the carrier gas flow rate ratio based on the result of the measurement.

5   Thus, in the case of using FTIR, it is possible to detect a concentration ratio of the source gases directly even in a low-pressure environment as used in an MOCVD process. On the other hand, it is not always easy to correct the

10   concentration of the source gases in such a process when it was found, as a result of the FTIR measurement, that the source gas concentration ratio is deviated from a proper concentration range.

        More specifically, the foregoing prior art

15   film-formation process compensates for the change of source gas concentration by increasing or decreasing the carrier gas flow rate when it was judged by the FTIR measurement that the source gas concentration is deviated from a proper concentration range.

20   In such a control scheme, there is a possibility that increase or decrease of the carrier gas flow rate invites, depending on the vaporization rate of the liquid or solid source material used and also on the carrier gas flow rate, an unpredictable

25   change of source gas concentration for the source gas that is actually introduced into the processing vessel.

        Consider now the case of increasing the carrier gas flow rate, under the situation that the

30   source gas concentration in the carrier gas is judged as being smaller than the proper concentration range, such that the vaporization of the source material is facilitated and the concentration of the source gas

- 6 -

supplied to the CVD film-formation chamber is
increased as a result. There can be a case in which
the vaporization of the source material cannot follow
the increase of the carrier gas flow rate. When this
5   is the case, the source gas produced from the source
material is merely diluted by a large amount of the
carrier gas, and the source gas concentration in the
carrier gas is reduced, contrary to what is intended.
Thereby, it becomes necessary to conduct a complex
10   and time-consuming control process for recovering the
desired source gas concentration.

          In the case of decreasing the carrier gas
flow rate under the situation in which the source gas
concentration in the carrier gas is judged as being
15   larger than the proper concentration range, so as to
reduce the vaporization of the source material and
decrease the concentration of the source gas
introduced into the CVD chamber, the source gas
vaporized from the source material is concentrated as
20   a result of use of small amount of carrier gas. When
this is the case, the source gas concentration in the
carrier gas is increased, contrary to what is
intended.

          Further, while it is possible to control
25   the source gas concentration by adjusting the
temperature of the liquid or solid source material,
such a procedure is not realistic for the exact and
quick control of the source gas concentration in view
of the fact that the vaporization rate changes
30   drastically with the bottle temperature and in view
of the fact that it requires a very rigorous
temperature regulation for achieving the desired gas
concentration. In addition, it should be noted that

- 7 -

the response speed of the source gas temperature is
generally slow even when the vaporization temperature
itself can be changed quickly and exactly during the
film-formation process. Thus, the use of such a
5   process of controlling the vaporization temperature
is not realistic in the situation in which there is a
demand of quick and exact adjustment of the source
gas concentration.

10   SUMMARY OF THE INVENTION
          Accordingly, it is a general object of the
present invention to provide a novel and useful film-
formation apparatus wherein the foregoing problems
are eliminated.
15   Another and more specific object of the
present invention is to provide a CVD apparatus as
well as a source gas supplying apparatus used
therefor, wherein the concentration of a source gas
supplied to a processing vessel of the CVD apparatus
20   together with a carrier gas for conducting a CVD
process is adjusted with high precision and with high
speed even when the CVD process is conducted by using
a low vapor pressure source material.
          Another object of the present invention is
25   to provide a film-formation apparatus, comprising:
          a film-formation chamber; and
          a source gas supplying apparatus supplying
a source gas to said film-formation chamber together
with a carrier gas,
30   said source gas supplying apparatus
comprising:
          a detector detecting a concentration of
said source gas; and

- 8 -

a gas flow controller controlling a flow rate of an inert gas added to said carrier gas based on a result of measurement of said concentration of said source gas obtained by said detector.

5   According to the present invention, it becomes possible to supply the source gas always with a proper concentration at the time of film formation process, by controlling the concentration of the source gas before or during the film-formation

10   process. As a result, it becomes possible to conduct the film formation process with excellent film quality, reliability and reproducibility. As the source gas is adjusted to have the proper concentration in the state the inert gas is added

15   thereto, any deviation from the proper concentration can be immediately corrected by merely increasing the flow rate of the inert gas, as in the case of when it is judged that the measured concentration of the source gas has exceeded the upper limit of the proper

20   compositional range. In the event the measured concentration of the source gas has decreased below the lower limit of the proper concentration range, on the other hand, the proper concentration is restored by merely decreasing the flow rate of the inert gas.

25   It should be noted that this inert gas functions as a diluting gas, and the present invention controls the flow rate of this diluting gas based on the source gas concentration measured by the detector with reference to the predetermined proper

30   concentration range. This adjustment of the source gas concentration by way of control of the inert gas added to the source gas is predictable and can be achieved with high precision and high speed, in

contrast to the case of attempting such an adjustment by way of controlling the carrier gas flow rate.

It should be noted that the concentration of the source gas is measured in the state the inert
5 gas is added thereto in the present invention. In other words, the concentration of the source gas is measured in the state it is introduced into the CVD apparatus for film formation. By measuring the concentration of the source gas in the flow passage
10 that is connected to the processing chamber of a CVD apparatus, and particularly by measuring the concentration of the gas directly, a direct and reliable control of the source gas concentration becomes possible.

15 By using a Fourier transform infrared spectrometer or a non-dispersion type infrared spectrometer, which shows high precision and high sensitivity also under a low-pressure environment as the means of the measurement of the source gas
20 concentration, it becomes possible to control the film formation process that uses a low vapor pressure source material such as a solid source material effectively. As noted before, there is a tendency that the source gas flow rate fluctuates
25 significantly in the case a solid source material is used.

Another object of the present invention is to provide a film-formation apparatus, comprising:

a film-formation chamber; and
30 a source gas supplying apparatus supplying a source gas to said film-formation chamber together with a carrier gas via a gas passage in the form of a mixed gas,

– 10 –

said source supplying apparatus comprising:

a gas concentration measurement part measuring the concentration of said source gas contained in said mixed gas in said gas passage;

5   a gas concentration controller connected to said gas passage, said gas concentration controller adding an inert gas to said mixed gas in said gas passage; and

an inert-gas flow-rate controller
10 controlling the flow rate of said inert gas added by said gas concentration controller based on a measured concentration of said source gas obtained by said gas concentration measurement part,

said gas concentration measurement part
15 including a manometer for measuring the pressure of said mixed gas in said gas passage, said gas concentration measurement part correcting said measured concentration of said source gas based on a pressure measured by said manometer.

20   Another object of the present invention is to provide a gas concentration detection method, comprising the steps of:

supplying a mixed gas containing therein a source gas to a flow passage;

25   measuring the pressure of said mixed gas in said flow passage;

injecting infrared light to said mixed gas in said flow passage;

acquiring an absorption spectrum of said
30 source gas by detecting said infrared light after said infrared light has passed through said mixed gas in said flow passage;

acquiring the concentration of said source

- 11 -

gas in said mixed gas by correcting an intensity of said absorption spectrum, said step of correction comprising the step of applying a correction term including therein said pressure.

5   According to the present invention, it becomes possible to obtain the absolute value of the source gas concentration by injecting a signal into the mixed gas that contains therein the source gas, detecting the signal after it has passed through the

10   mixed gas, and correcting the detected signal by using a correction factor that contains therein the term of total pressure of the mixed gas.

Other objects and further features of the present invention will become apparent from the

15   following detailed description when read in conjunction with the attached drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG.1 is a diagram showing the construction

20   of a processing vessel of an MOCVD apparatus used in the present invention;

FIG.2 is a diagram showing the construction of an MOCVD apparatus according to a first embodiment of the present invention;

25   FIG.3 is a diagram showing the construction of an MOCVD apparatus according to a second embodiment of the present invention;

FIG.4 is a diagram showing the construction of an MOCVD apparatus according to a third embodiment

30   of the present invention;

FIG.5 is a diagram showing the construction of an MOCVD apparatus according to a fourth embodiment of the present invention;

- 12 -

FIG.6 is a flowchart showing an example of processing for controlling a source gas concentration in a mixed gas according to any of the first through fourth embodiment;

5   FIG.7 is a diagram showing an example of an FTIR spectrum obtained for $W(CO)_6$;

FIG.8 is a diagram showing the construction of an MOCVD apparatus according to a fifth embodiment of the present invention;

10   FIG.9 is a diagram showing the construction of an MOCVD apparatus according to a modification of the MOCVD apparatus of FIG.8;

FIG.10 is a diagram showing the construction of an MOCVD apparatus according to

15   another modification of the MOCVD apparatus of FIG.8;

FIG.11 is a diagram showing the construction of an MOCVD apparatus according to a further modification of the MOCVD apparatus of FIG.8;

FIG.12 is a diagram showing the

20   construction of an MOCVD apparatus according to a further modification of the MOCVD apparatus of FIG.8;

FIG.13 is an MOCVD apparatus according to a further modification of the MOCVD apparatus of FIG.8;

FIG.14 is a diagram showing the

25   construction of an FTIR apparatus according to a sixth embodiment of the present invention;

FIG.15 is a diagram showing the construction of a non-dispersion infrared spectrometer according to the sixth embodiment of the

30   present invention; and

FIG.16 is a diagram showing a further modification of the MOCVD apparatus of FIG.12.

- 13 -

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[FIRST EMBODIMENT]

FIG.1 shows the construction of a
processing vessel 100 used in a first embodiment of
5   the present invention in a cross-sectional view.

Referring to FIG.1, the processing vessel
100 includes a processing vessel body 120 and a stage
130 provided in the processing vessel body 120 for
supporting a semiconductor wafer 101, wherein the
10   stage 130 is embedded with a heating element 132
driven by a power source 132A, and there is provided
a shower head 110 inside the processing vessel body
120 so as to face the stage 130. The showerhead 110
introduces a gas supplied from a source gas line 30
15   into the process space inside the processing vessel
body 120.

Further, there is provided a gate valve 140
at the sidewall of the processing vessel body 120 for
loading and unloading the semiconductor wafer 101 to
20   and from the processing vessel body 120. The
processing vessel body 120 is evacuated via an
evacuation line 32.

FIG.2 shows the construction of an MOCVD
apparatus 200 that uses the processing vessel 100 of
25   FIG.1 schematically.

Referring to FIG.2, the MOCVD apparatus 200
includes a source bottle 10, wherein the source
bottle 10 is supplied with an inert gas such as Ar,
Kr, N_2, H_2, and the like, from a source gas line 30
30   via a mass flow controller (MFC) 12A, which is
provided in a part of the source gas line 30. Thereby,
the mass-flow controller 12A controls the flow rate
of the inert gas supplied to the source bottle 10.

- 14 -

The source bottle 10 accommodates therein a liquid or solid source material and produces a source gas therein as a result of vaporization of the source material. The inert gas thus supplied to the source
5   bottle 10 functions as a carrier gas and transports the source gas from the source bottle 10 to the processing vessel 100. Thereby, the source gas flows out from the source bottle 10 at the outlet port thereof and is transported along the source gas line
10   30. It should be noted that there is provided a manometer 18 in the vicinity of the outlet port of the source bottle 10 for detecting the pressure inside the source bottle 10.

In the MOCVD apparatus 200 of FIG.14, it
15   should further be noted that there is provided a diluting gas line 31 to the source gas line 30 so as to merge at a downstream side of the manometer 18, and an inert gas such as Ar, Kr, N_2, H_2, and the like, is supplied to the diluting gas line 31 via another
20   mass-flow controller 12B. It should be noted that the mass-flow controller 12B controls the flow rate of the inert gas added to the source gas line 30.

It should be noted that this inert gas in the diluting gas line 31 functions as a diluting gas
25   when it is added to the source gas line 30 and dilutes the source gas transported through the source gas line 30 from the source bottle 10. Hereinafter, the gas in the source gas line 30 thus diluted by the inert gas in the gas line 31 will be referred to as a
30   "mixed gas". This mixed gas is supplied to the processing vessel 100 through the source gas line 30.

In the construction of FIG.2, a turbo molecular pump (TMP) 14 is provided to the evacuation

- 15 -

line 32 connected to the processing vessel 100, and
there is further provided a dry pump (DP) behind the
turbo molecular pump 14 for boosting the same. By
driving these pumps 14 and 16, the interior of the
5   processing vessel body 120 is maintained at a
predetermined pressure. For example, the turbo
molecular pump 14 can evacuate the process space
inside the processing vessel 120 to a high-degree
vacuum state characterized by the pressure of about 1
10   Torr (133Pa). Thereby, it becomes possible to conduct
the film formation process that uses a source
material of low vapor pressure.

It should be noted that there is provided a
pre-flow line 33 in the source gas line 30 so as to
15   bypass the processing vessel 100 at the downstream
side of the source bottle 10, and the mixed gas in
the source gas line 30 is supplied selectively to one
of the pre-flow line 33 or to the source gas line 30
connected to the processing vessel 100 by the
20   switching of a valve 26 provided on the source gas
line 30 from the source bottle 10. The pre-flow line
33 is provided for stabilizing the flow rate of the
mixed gas supplied to the processing vessel 100 at
the time of the film-formation process and to pre-
25   adjust the concentration of the mixed gas. Thus, the
mixed gas is caused to flow through the pre-flow line
33 in advance to each step of processing the
substrate 101.

Meanwhile, the mixed gas supplied to the
30   processing vessel 100 has to contain the source gas
with a proper concentration range in order to achieve
the desired film formation. Further, the mixed gas is
required to contain the source gas with a constant

- 16 -

concentration within the proper concentration range
in order to avoid variation of film quality in each
film-formation step when the film-formation step is
conducted repeatedly.

5   In the MOCVD apparatus 200 of FIG.2, on the
other hand, there arises a difficulty of detecting
the source gas concentration accurately because of
the fact that the carrier gas and also the diluting
gas are added to the source gas when supplying the
10   source gas to the processing vessel 100, contrary to
the case of supplying the source gas directly from a
vaporizer. As noted previously, the concentration of
the source gas tends to change with the pressure
variation or variation of the surface area of the
15   source material, particularly when a solid source
material is used, and it is difficult stabilize the
concentration of the source gas in the mixed gas.
          Thus, according to a first embodiment of
the present invention, variation of the source gas
20   concentration in the mixed gas is detected with high
precision by using a Fourier-transform infrared
spectrometer, and the flow rate of the inert gas is
controlled by using the mass-flow controller 12B
and/or 12A such that the source gas concentration in
25   the mixed gas falls always in a predetermined, proper
concentration range.
          More specifically, the present embodiment
provides a Fourier-transform infrared spectrometer 40
referred to hereinafter as FTIR 40 in the pre-flow
30   line 33 of the MOCVD apparatus 200, wherein the FTIR
40 includes a wave monitor using a laser light and a
movable mirror. More specifically, the FTIR 40
includes an interferometer, infrared detector and a

- 17 -

processing unit and measures the concentration of gas
species contained in a gas based upon the absorption
spectrum of respective gas species, by irradiating an
infrared beam upon the gas via an interferometer and
5   processing the output of the infrared detector by the
processing unit.

The pre-flow line 33 is merged to the
evacuation line 32 at the upstream side of the dry
pump 16 and is set to the predetermined degree of
10   vacuum by the dry pump 16. In the MOCVD apparatus 200
of FIG.2, it becomes possible to measure the source
gas concentration under the pressure of 50 Torr
(6660Pa) or less, in which concentration measurement
by way of acoustic emission process is not possible,
15   by providing the FTIR 40 in the pre-flow line 33.

In the present embodiment, it should be
noted that the FTIR 40 provided in the pre-flow line
33 measures the source gas concentration in the mixed
gas (referred to hereinafter as "measured
20   concentration") and supplies a signal indicative of
the measured concentration to a controller 201.

Thus, in the event it is judged in the
controller 201 that the measured concentration change
has exceeded a predetermined range, the controller
25   201 controls the mass-flow controller 12B and/or 12A,
and increases or decreases the flow rate of the inert
gas. It should be noted that this controller 201 may
be provided inside the FTIR 40 or inside any of the
mass-flow controller 12B and/or 12A.

30   According to the first embodiment of the
present invention, the source gas concentration in
the mixed gas is controlled constant before
conducting a film formation step for each substrate,

- 18 -

and the source gas is introduced into the processing
vessel with controlled concentration in each film-
formation process. Thereby, wafer-to-wafer variation
of film quality is successfully minimized.

5   Because the source gas concentration is
adjusted to the proper concentration range at the
beginning of the process by adding the diluting gas
to the carrier gas, it is possible to increase the
source gas concentration immediately, in the event

10   the source gas concentration has decreased during the
film-formation process as a result of decrease of the
source material or as a result of decrease of
vaporization efficiency of the source material, and
the like, by decreasing the flow rate of the diluting

15   gas. Similarly, it is possible to decrease the source
gas concentration immediately in the event the source
gas concentration has increased during the film-
formation process, by increasing the flow rate of the
diluting gas.

20   As explained before, increase of the
carrier gas flow rate does not always lead to
increase of source gas concentration. This is
particularly true in the case of using a solid source
material, in which there is caused a reduction of

25   surface area with the progress of the film-formation
process. In such a case, the efficiency of
vaporization decreases when the carrier gas flow rate
is increased.

          In the present invention, on the other hand,
30   it is possible to increase the source gas
concentration by merely decreasing the flow rate of
the diluting gas. Of course, it is possible to change
the carrier gas flow rate simultaneously to the

- 19 -

change of the diluting gas flow rate.

As the diluting gas does not contain the source gas, it is easy to determine the amount of the flow rate change from the measured concentration
5   obtained by FTIR and from the predetermined proper concentration range. Thus, the concentration control of the source gas by the increase or decrease of the diluting gas can be conducted at high speed with high precision, contrary to the case of conducting the
10   concentration control by way of control of the carrier gas alone.

[SECOND EMBODIMENT]

FIG.3 shows the construction of an MOCVD
15   apparatus 200A according to a second embodiment of the present invention, wherein those parts corresponding to the parts described previously are designated by the same reference numerals and the description thereof will be omitted.
20   Referring to FIG.3, it can be seen that the inert gas such as Ar, Kr, N_2, H_2, and the like, is supplied to the source bottle 10 via the source gas line 30 via the mass-flow controller 12A, wherein the mass-flow controller 12A controls the flow rate of
25   the inert gas to be supplied to the source bottle 10. The source bottle 10 accommodates therein a liquid or solid source used for the film-formation process. Thereby, the source gas is produced as a result of the vaporization of the source material in the source
30   bottle 10. The inert gas thus supplied to the source bottle 10 is forwarded to the source gas line 30 from the outlet port of the source bottle 10 as the carrier gas.

- 20 -

        According to the second embodiment of the
present invention, it can be seen that the diluting
gas line 31 is provided to the source gas line 30 at
the downstream side of the mass-flow controller 12A

5   so as to bypass the source bottle 10, and the
diluting gas line 31 is provided with the inert gas
diverted from the source gas line 30. This inert gas
is admixed to the carrier gas carrying the source gas
from the source bottle 10 upon merging to the source

10   gas line at the node B as the diluting gas, and the
mixed gas formed of the source gas, carrier gas and
the diluting gas is caused to flow through the source
gas line 30 at the downstream side of the node B.
Thereby, the source gas in the mixed gas is diluted

15   by the inert diluting gas in the gas line 31. The
flow rate of this diluting gas is controlled by a
valve 20 provided in the gas line 31.

        This mixed gas is then supplied selectively
to one of the processing vessel 100 or the pre-flow

20   line 33 in which the FTIR 40 is provided, after
passing through the source gas line 30.

        The FTIR 40 of the pre-flow line 33
measures the source concentration in the mixed gas
and supplies an output signal indicative of the

25   measured concentration to the controller 201. Thus,
the controller 201 judges whether the measured
concentration falls within the predetermined proper
concentration range or not and conducts the control
of increasing or decreasing the diluting gas flow

30   rage by controlling the valve 20 when it is judged
that the measured concentration deviated beyond the
proper concentration range.

        According to the second embodiment of the

- 21 -

present invention, it becomes possible to minimize
the wafer-to-wafer variation of the film quality by
monitoring the source gas concentration by using the
FTIR 40 provided in the pre-flow line 33, similarly

5   to the first embodiment. Further, the present
embodiment, which uses FTIR for the concentration
measurement, is suitable for the film-formation
process that uses the source material of low vapor
pressure. Further, any deviation of the source gas

10   concentration in the mixed gas beyond the proper
concentration range is corrected immediately by
increasing or decreasing the flow rate of the
diluting gas.

       In the present embodiment in which the flow

15   rate of the diluting gas is controlled by the valve
20 provided in the diluting gas line 31, it is
possible to adjust the flow rate of the diluting gas
and the carrier gas by using the single mass-flow
controller 12A. Further, the construction of the

20   present embodiment, in which the diluting gas line 31
is branched from the line 30 and merged thereto again,
has the feature that the inert gas flow rate before
branching becomes generally equal to the inert gas
flow rate at the merging node B. Thus, it becomes

25   possible to maintain a constant flow rate for the
mixed gas supplied to the processing vessel 100 while
controlling the source gas concentration therein by
way of increasing or decreasing the diluting gas flow
rate. As a result, wafer-to-wafer variation of film

30   quality is reduced further by using the present
embodiment.

[THIRD EMBODIMENT]

- 22 -

FIG.4 shows the construction of an MOCVD apparatus 200B according to a third embodiment of the present invention, wherein those parts corresponding to the parts described previously are designated by
5　the same reference numerals and the description thereof will be omitted.

Referring to FIG.4, it can be seen that the inert gas such as Ar, Kr, N_2, H_2, and the like, is supplied to the source bottle 10 from the mass-flow
10　controller 12A through the source gas line 30, wherein the mass-flow controller 12A controls the flow rate of the inert gas supplied to the source bottle 10. The source bottle 10 accommodates therein a liquid or solid source material used for film-
15　formation, and the source material undergoes vaporization in the source bottle 10. The inert gas thus supplied to the source bottle 10 functions as a carrier gas and carries the source gas therewith. Further, the manometer 18 is provided in the vicinity
20　of the gas outlet of the source bottle 10 for detecting the pressure in the source bottle 10.

The source gas line 30 is further provided with the diluting gas line 31 such that the diluting gas line 31 merges at the downstream side of the
25　manometer 18, and the diluting gas line 31 is supplied with an inert gas such as Ar, Kr, N_2, H_2, and the like, via the mass-flow controller 12B. Thereby, mass-flow controller 12B controls the flow rate of the inert gas to be merged to the source gas
30　line 30. It should be noted that this inert gas is admixed to the source gas and the carrier gas from the source bottle 10 upon merging to the source gas line 30, and the mixed gas is formed in the source

- 23 -

gas line 30 as a result of the admixing. Thereby, the inert gas thus added causes dilution of the source gas contained in the mixed gas. The mixed gas thus formed is then supplied to the processing vessel 100

5 through the source gas line 30. In the present embodiment, it is also possible to construct the diluting gas line 31 similar to the one explained with reference to the second embodiment.

In the present embodiment, the evacuation

10 line 32 for evacuating the processing vessel 100 is provided with the turbo molecular pump 14, and the turbo molecular pump 14 is boosted by the dry pump 16 provided at the downstream side of the turbo molecular pump 14. Thereby, the process space in the

15 processing vessel 100 is maintained at a predetermined pressure or predetermined degree of vacuum such as 1 Torr (133Pa) or less. Such a low pressure environment is particularly important in the film-formation process that uses a low vapor pressure

20 source material.

It should be noted that the source gas line 30 is provided with the pre-flow line 33 such that the pre-flow line 22 bypasses the processing vessel 100, wherein the pre-flow line 33 is supplied with

25 the mixed gas in the source gas line 30. Thereby, the mixed gas is supplied selectively to one of the pre-flow line 33 and the source gas line 30 connected to the processing vessel 100 by the activation of the valve 26. Here, the pre-flow line 33 is provided for

30 stabilizing the flow rage of the mixed gas supplied to the processing vessel 100 at the time of the film-formation process and to adjust the concentration of the mixed gas in advance. As can be seen, this pre-

- 24 -

flow line 33 merges to the evacuation line 32 at the
upstream side of the dry pump 16. Thus, the pressure
inside the pre-flow line 33 is determined by the dry
pump 16.

5   In the first and second embodiments
described before, it should be noted that the
concentration of the mixed gas supplied to the
processing vessel 100 is monitored by the FTIR 40
provided in the pre-flow line 33. In the case of

10   switching the mixed gas hitherto supplied to the pre-
flow line 33, to the source gas line 30 connected to
the processing vessel in the gas supply system having
such a construction, there can be a case that the
source gas concentration undergoes a change before

15   and after the switching as a result of different
diameter of the gas lines or existence or non-
existence of the processing vessel body 120 in the
evacuation path, which causes a change of impedance
in the evacuation system, and such a change of

20   impedance of the evacuation system can cause a change
of pressure in the source bottle 10. In the case of
conducting film-formation by using a low vapor
pressure source, in particular, the pressure inside
the source bottle 10 is held at the pressure of 1

25   Torr (133Pa) or less because of the use of the turbo
molecular pump 14 for facilitating the vaporization
of the source material in the source bottle 10. On
the other hand, the pre-flow line 33 is evacuated by
the dry pump 16 alone, and thus, it is difficult to

30   realize such a low pressure in the pre-flow line 33.
        Even in such a case, the first and second
embodiments can successfully reduce the wafer-to-
wafer variation of film quality by maintaining the

source gas concentration in the mixed gas flowing through the pre-flow line 22 before each film-formation step.

However, it would be more advantageous when
5　a direct control of the source gas concentration in the mixed gas supplied actually to the processing vessel is achieved. Thereby, it should be noted that such a direct control of the source gas concentration in the mixed gas actually supplied to the processing
10　vessel 100 has to be conducted in a short time period in view of the fact that the film-formation process is in progress in the processing vessel 100 while using this mixed gas.

The present invention can immediately
15　correct any deviation of the source gas concentration by way of increase of decrease of the diluting gas, and thus, it becomes possible to control the source gas concentration in the mixed gas supplied directly to the processing vessel 100.

20　More specifically, the present embodiment provides the FTIR 40 in the source gas line 30 connected to the processing vessel 100 for measuring the source gas concentration of the mixed gas introduced into the processing vessel 100. It should
25　be noted that this source gas line 30 may be held at a low pressure such as 1 Torr (133Pa) or less by using the turbo molecular pump 14 and the dry pump 16 in order to facilitate vaporization of the low vapor pressure source material. Even in such a case, the
30　source gas concentration is measured accurately by using the FTIR 40.

In the present embodiment, the FTIR 40 provided in the source supply line 30 connected to

- 26 -

the processing vessel 100 measures the source gas
concentration in the mixed gas and supplied an output
signal indicative of the measured concentration to
the controller 201. When the controller 201 judges
5   that the measured concentration of the FTIR 40 has
exceeded the proper concentration range, the
controller 201 controls the mass-flow controller 12B
and/or 12A and the flow rate of the inert gas added
to the mixed gas is increased or decreased.
10   According to the fourth embodiment of the
present invention, it becomes possible to immediately
correct any deviation of the source gas concentration
in the mixed gas from the proper concentration range
by increasing or decreasing the diluting gas flow
15   rate similarly to the previous embodiments.
             Thus, the present embodiment enables direct
measurement of the source gas concentration in the
mixed gas, which is actually used for film formation,
by using the FTIR 40 provided in the source gas line
20   40 connected to the processing vessel 100. Thereby,
any deviation of the source gas concentration in the
mixed gas currently in use in an on-going film
formation process is detected and corrected
immediately. Thereby, the film-formation processing
25   can be conducted by using a source gas of which
concentration is always controlled to the proper
compositional range, and the desired film quality is
maintained over the repeatedly conducted wafer-to-
wafer film-formation processes.
30

[FOURTH EMBODIMENT]
             FIG.5 shows the construction of an MOCVD
apparatus 200C according to a fourth embodiment of

the present invention, wherein those parts
corresponding to the parts described previously are
designated by the same reference numerals and the
description thereof will be omitted.

5   Referring to FIG.5, the inert gas such as
Ar, Kr, N_2, H_2, and the like, is supplied to the
source bottle 10 from the mass-flow controller 12A
through the source gas line 30, wherein the mass-flow
controller 12A controls the flow rate of the inert
10   gas supplied to the source bottle 10. The source
bottle 10 accommodates therein a liquid or solid
source material used for the film-formation process,
and the source gas is produced as a result of
vaporization conducted in the source bottle 10.
15   Thereby, the inert gas supplied to the source bottle
10 functions as a carrier gas and carries the
vaporized source gas therewith. Further, the
manometer 18 is provided in the vicinity of the
outlet port of the source bottle 10 connected to the
20   source gas line 30 for detecting the pressure inside
the source bottle 10.

            Further, there is provided the diluting gas
line 31 such that the diluting gas line 31 merges the
source gas line 30 at the downstream side of the
25   manometer 18, and the inert gas such as Ar, Kr, N_2,
H_2, and the like, is supplied to the diluting gas
line 31 via the mass-flow controller 12B. Thereby,
the mass-flow controller 12B controls the flow rate
of the inert gas added to the source gas supply line
30   30. It should be noted that this inert gas is added,
upon merging to the source gas line 30, to the source
gas and the carrier gas from the source bottle 10 as
a diluting gas, and there is formed a mixed gas in

- 28 -

the source gas line 30 as a result of the mixing of
the source gas, carrier gas and the diluting gas at
the downstream side of the node where the gas line 31
merges the gas line 30. This mixed gas is then

5   supplied to the processing vessel 100 along the
source gas supply line 30. Here, it is possible to
construct the diluting gas line 31 and the source gas
supply line 30 similarly to the second embodiment.

The source gas supply line 30 is provided

10   with the pre-flow line 33 so as to bypass the
processing vessel 100 at the downstream side of the
source bottle 10, and the pre-flow line 33 is
supplied with the mixed gas from the source gas line
30. The mixed gas is thereby supplied selectively to

15   one of the pre-flow line 33 and the source gas line
30 connected to the processing vessel 100 by the
activation of the valve 26. It should be noted that
this pre-flow line 33 is provided so as to stabilize
the flow rate of the mixed gas supplied to the

20   processing vessel 100 at the time of the film
formation process and further to adjust the mixed gas
concentration in advance. The pre-flow line 33 is
connected to the evacuation line 32 at the upstream
side of the dry pump 16. Thus, the pre-flow line 33

25   is evacuated to a predetermined pressure or degree of
vacuum by the dry pump 16.

According to the fourth embodiment of the
present invention, the FTIR 40 is provided at the
downstream side of the node where the gas line 31

30   merges the gas line 30 but at the upstream side of
the node where the pre-flow line 33 branches from the
gas line 30, so as to enable measurement of the
source gas concentration in the mixed gas introduced

- 29 -

into the processing vessel 100 and so as to enable
measurement of the source gas concentration in the
mixed gas flowing through the pre-flow line 33.

As shown in FIG.5, the FTIR 40 may be
5 provided in a bypass line 35 bypassing from the
source gas line 30. The bypass line 35 is provided
with valves 21 and 25, and there is further provided
a valve 23 in the source gas line 30 between the node
where the bypass line 35 branches from the source gas
10 line 30 and the node where the bypass line 35 merges
the source gas line 30.

As a result of opening and closing of these
valves 21, 23 and 25, the mixed gas is supplied
selectively to either the bypass line 35 or the
15 source gas line 30. Thus, the mixed gas is supplied
to the bypass line 35 the source gas concentration is
to be measured. When there is no need of measurement
of the source gas concentration, the source gas is
supplied directly to the downstream part of the
20 source gas line 30.

The output of the FTIR 40 is supplied to
the controller 201, and the controller 201 controls
the mass-flow controller 12A and/or 12B in response
to the output signal of the FTIR 40.

25 According to the fourth embodiment
described above, it is possible to correct any
deviation of the source gas concentration in the
mixed gas beyond a predetermined proper concentration
range immediately by increasing or decreasing the
30 flow rate of the diluting gas.

As the FTIR 40 is disposed so as to measure
both the source gas concentration introduced into the
processing vessel and the source gas concentration in

- 30 -

the pre-flow line 33, the present embodiment can
control not only the source gas concentration before
the commencement of the film-formation process but
also the source gas concentration actually used for
5   the film-formation process. In other words, the
present embodiment enables monitoring of the source
gas concentration of the mixed gas actually in use
for the film formation process and correction of any
deviation of the source gas concentration beyond the
10   predetermined concentration range immediately. As the
mixed gas used for the actual film-formation process
has the source gas concentration already adjusted
during the period in which the mixed gas is caused to
flow through the pre-flow line, there can occur no
15   large deviation in the source gas concentration when
the mixed gas is switched and introduced into the
processing vessel 100. Thus, the need of large change
of the source gas concentration during the film-
formation process by significantly increasing or
20   decreasing the diluting gas is avoided, and a stable
film formation becomes possible.

        While the present invention has been
described for the case of conducting film-formation
process by using a single gas source specie, the
25   present invention is applicable also to the case of
conducting film-formation processing by using two or
more gas species. In such a case, two or more source
gas lines are provided for supplying the respective
source gases to the CVD apparatus. Otherwise, the CVD
30   apparatus has a construction similar to those
described before.

        It should be noted that the pre-flow line
33 is indicated as merging to the evacuation lien 32

- 31 -

at the upstream side of the dry pump 16. In such a
case, there is a possibility that the source gas
concentration is measured in the state in which the
pressure of the mixed gas is higher than the case in
5   which the mixed gas actually flows to the processing
vessel 100. In order to avoid this problem and to
measure the source gas concentration at the pressure
in which the mixed gas is actually supplied to the
processing vessel 100, it is possible to increase the
10   diameter of the pipe of the pre-flow line 33 for the
part extending from the outlet of the FTIR to the
node where the pre-flow line 33 merges the evacuation
line 32 at the upstream side of the dry pump 16.
Alternatively, it is possible to merge the pre-flow
15   line 33 to the evacuation line 32 not at the upstream
side of the dry pump 16 but at the upstream side of
the turbo molecular pump 14. Thereby, it is possible
to provide a pressure regulation valve not
illustrated in the pre-flow line 33 at the upstream
20   side of the node where the pre-flow line 33 merges
the evacuation line 32 and control the cell pressure
to the pressure of the film-formation process when
the FTIR 40 is activated to conduct the measurement
of the source gas concentration.
25   Next, the control operation of the
controller 201 in the various embodiments described
heretofore will be explained.
            FIG.6 shows an embodiment of control
routine for controlling the source gas concentration
30   in the mixed gas. While the illustration is omitted,
this controller 201 is formed of a microcomputer
including a CPU as a major component and stores the
target source gas concentration C1 of the mixed gas,

- 32 -

initial designed flow rate value Q1 of the diluting gas, the initial designed flow rate value Q2 of the carrier gas, and the like, in a memory.

In the step S300, the microcomputer sets
5   the flow rate of the diluting gas to the value Q1 based on the data stored in the memory and produces a control signal setting the carrier gas flow rate to the value Q2. This control signal is then supplied to the mass-flow controllers 12A and 12B.

10   Next, in the step 302, the microcomputer determines the flow rate of the diluting gas and the flow rate of the carrier gas in response to the measured concentration C2 supplied from the FTIR 40 and determines the flow rate of the diluting gas and
15   the flow rate of the carrier gas such that the measured concentration C2 coincides with the target concentration C1. Alternatively, the microcomputer decides whether or not the measured concentration C2 is deviated from the target concentration C1 beyond
20   the allowable range. In this case, the microcomputer determines the flow rate of the diluting gas and the flow rate of the carrier gas such that the measured concentration C2 falls in the allowable range only when it is judges that the measured concentration C2
25   has deviated beyond the foregoing allowable range.

In the present embodiment, the overall flow rate of the diluting gas and the carrier gas is set constant before and after the adjustment. Thus, the flow rate of the diluting gas after the adjustment
30   can be represented as $Q1'=Q1+\beta$ and the flow rate of the carrier gas after the adjustment can be represented as $Q2'=Q2-\beta$. In the adjustment, the parameter β is determined.

- 33 -

Thus, in the initial routine step 302, the term $-Q2/10$ or $+Q2/10$ is substituted into the parameter β and the initial values Q1 and Q2 are updated to Q1' and Q2'. The values thus updated are

5   stored in the memory, and the corresponding control signals are transmitted to the mass-flow controllers 12A and 12B. Further, the step 302 is repeated in response to the input from the FTIR 40.

In the step 302 of the next routine, a new

10   parameter β is determined in such a manner that the difference between the newly measured concentration C2 and the target concentration C1 is decreased and such that the newly determined parameter β has a smaller magnitude. By using the newly determined

15   parameter β, the initial values Q1 and Q2 are updated to Q1' and Q2' and stored in the memory. Thereby, the corresponding control signals are transmitted to the mass-flow controllers 12A and 12B.

20   [EXAMPLE 1]

FIG.7 shows an example of the infrared absorption spectrum of the metal organic gas $W(CO)_6$ (hexacarbonyl tungsten), wherein the horizontal axis represents the wavenumber while the vertical axis

25   represents the transmissivity.

From FIG.7, it can be seen that there exits characteristic absorption of carbonyl group (=CO) in the metal organic gas $W(CO)_6$ at the wavenumers of about $2900cm^{-1}$, $1900cm-1$ and $500cm^{-1}$.

30   In order to confirm the sensitivity of the FTIR 40 for the concentration change of the $W(CO)_6$ gas, an experiment was made in which the source bottle 10 is held at the temperatures of 25℃, 45℃

- 34 -

and 60℃ and an Ar carrier gas is supplied with the
flow rate of 50 SCCM. Thereby, the FTIR 40 was
provided not in the pre-flow line 33 but at the
upstream side of the processing vessel 100 as in the

5 case of the third embodiment. Here, the pressure of
the FTIR cell becomes 80Pa, 85Pa and 87Pa,
respectively, and it was found that the corrected
absorbance of the carbonyl group corrected to the
pressure of 1330Pa (10 Torr) is 0.337, 0.656 and

10 1.050, respectively. From this result, it was
confirmed that the FTIR has a sufficient sensitivity
even at such low pressures and that it can be used
for the monitoring of the concentration change of the
W(CO)$_6$ gas by monitoring the change of the absorption

15 peak intensity.

[EXAMPLE 2]

        In Example 2, W(CO)$_6$ is used for the source
material and the source bottle 10 is held at 45℃.

20 Further, an Ar gas was used for the carrier gas and
the diluting gas. The FTIR 40 was provided at the
upstream side of the processing vessel 100, similarly
to the fourth embodiment, and the temperature of the
source bottle was set to 45℃. Thereby, the carrier

25 gas was supplied with the flow rate of 50 SCCM and
the diluting gas was supplied with the flow rate of
10 SCCM.

        In this experiment, a value of 0.235 was
obtained initially for the absorbance of W(CO)$_6$ as

30 corrected to the pressure of 1330Pa (10Torr) from the
absorption peak of the carbonyl group.

        After 5 minutes, it was found that the
value of the absorbance has changed to 0.267, and

- 35 -

thus, the flow rate of the diluting gas was increased slightly to the value of 12SCCM. With this, it became possible to change the absorbance to 0.233, which is close to the original value.

5

[EXAMPLE 3]

In Example 3, a tungsten film was formed by a pyrolytic CVD process while using $W(CO)_6$ as a source material.

10   In this experiment, the source bottle 10 was held at 60℃ and an Ar carrier gas was supplied with the flow rate of 300SCCM. Further, an Ar diluting gas was supplied with the flow rate of 100SCCM.

15   Further, in order to facilitate the vaporization of $W(CO)_6$, which provides only the vapor pressure of about 106Pa at 60℃, and to increase the film-formation rate, the present embodiment activates the turbo molecular pump 14 and the dry pump 16 such

20   that a process pressure of 0.15 Torr (about 20Pa) is realized in the processing vessel body 120 and the pressure of 1.5 Torr (about 200Pa) is realized in the source gas line 30.

As a result of the film formation conducted

25   at the substrate temperature of ·450℃, it was confirmed that there occurs a tungsten film formation with the rate of 7.1nm/min. The tungsten film thus formed had the resistivity of 25μΩcm.

30   [FIFTH EMBODIMENT]

As a result of various embodiments described heretofore, it became possible to maintain the concentration of the source gas supplied to the

processing vessel body 120 constant after one process has been commenced, by using FTIR 40 and the controller 201.

5 On the other hand, the embodiments explained above do not measure the absolute concentration of the source gas, and because of this, there has been a need, in the event one process has been terminated and the supply of the source gas is interrupted, to seek for the optimum condition of

10 film-formation upon commencement of the next film-formation process, by processing a number of test substrates so that the desired source gas concentration is attained. However, such a search of the optimum condition is time consuming and increases

15 the cost of the produced semiconductor device.

FIG.8 shows the construction of an MOCVD apparatus 200D according to a fifth embodiment of the present invention that is capable of measuring the absolute source gas concentration by using the FTIR

20 40, wherein those parts described previously are designated by the same reference numerals and the description thereof will be omitted.

Referring to FIG.8, the MOCVD apparatus 200D has a construction similar to the MOCVD

25 apparatus 200A explained before, except that there is provided another manometer 18A at the downstream side of a node P1 where the diluting gas line 31 merges the source gas line 30 but at the upstream side of the processing vessel 100, for measuring the pressure

30 of the mixed gas of the gas line 30 in the state that the Ar gas in the diluting gas line 31 is added to the gas in the source gas line 30.

The manometer 18A supplied the output

- 37 -

signal corresponding to the detected pressure to the
controller 201, and the controller 201 obtains the
absolute concentration of the source gas in the mixed
gas supplied to the processing vessel 100 via the
5   source gas line 30 based on the output of the FTIR 40
and the output of the manometer 18A.

In the construction that supplies a source
gas to the processing vessel 100 together with a
carrier gas along the gas line 30, there holds a
10   relationship

$$S = A \times Ir \times (1/P) \times C \qquad (1)$$

wherein A is a constant depending on a cell length, S
represents the flow rate of the source gas supplied
to the processing vessel 100, Ir represents the
15   absorption intensity of the source gas component in
the mixed gas transported through the source gas line
30 obtained by the FTIR 40, P represents the pressure
in the source gas line 30 connected to the processing
vessel 100, and C represents the total flow rate of
20   the carrier gas and the diluting gas in the gas line
30.

Thus, when the pressure P and the
carrier/diluting gas total flow rate C are held
constant and the source gas flow rate S is increased,
25   there occurs an increase of the output signal Ir of
the FTIR 40, while when the source gas flow rate S
and the output Ir of the FTIR 40 are held constant
and the pressure P is increased, there occurs an
increase of the total gas flow rate C. Further, when
30   the source gas flow rate increases with increase of
the carrier/diluting gas total flow rate C even when
the output Ir of the FTIR 40 and the pressure P are
held constant.

- 38 -

The foregoing Equation (1) can be modified
to

$$S/C = A \times Ir \times (1/P) \hspace{3cm} (2)$$

wherein it should be noted that the left side term

5   S/C is nothing but the absolute concentration of the
source gas in the mixed gas introduced into the
processing vessel 100.

Thus, Equation (2) means that it is
possible to calculate the absolute concentration of

10   the source gas in the mixed gas introduced into the
processing vessel 100 when the measurement of the
pressure P by the manometer 18A and the measurement
of the absorbance Ir by the FTIR 40 are both
conducted at the downstream side of the merging node

15   P1, by using the absorbance Ir and the pressure P.

In the case of using an FTIR having a cell
length of 1m for the device 40, the measurement
conducted under the atmospheric pressure (101.08kPa)
has yielded the data sets for the concentration S/C

20   and the infrared absorption intensity Ir, or (S/C,
Ir), of (0.314%, 1.19) and (0.043%, 0.21). From these
data sets, the foregoing constant A is obtained as
26.9 (A=26.9) in the case the pressure is represented
in terms of kilo Pascal.

25   Further, in the case of using an NDIR (non-
dispersion type infrared spectrometer) to be
described later, the data set for the pressure P and
the absorption intensity Ir, or (P, Ir), of (0.04kPa
(=0.3Torr), 0.0062) has been obtained for the NDIR

30   having a cell length of 1m, for the case the
measurement is conducted under the carrier gas flow
rate of 20 SCCM. In the case the measurement is
conducted with the carrier gas flow rate of 500 SCCM,

- 39 -

on the other hand, the data set of (0.20kPa
(=1.5Torr), 0.0008) has been obtained. From these
data sets, the source gas flow rate S is calculated
as 0.88SCCM and 0.56 SCCM, respectively, by using the
5   constant A of 26.9 obtained before.

It should be noted that the value of the
constant A depends on the cell length and becomes
1/10 in the case the cell length becomes ten times
larger.

10   Thus, by using the absolute concentration
in the step 302 of the flowchart of FIG.6 for
controlling the mass-flow controllers 12A and 12B by
way of the controller 201, it becomes possible to
control the absolute concentration of the source gas
15   to the predetermined desired value. This means that
it becomes possible to reproduce the optimum
deposition condition with reliability even in the
case a new film-formation process is restarted by
supplying the source gas after termination of a
20   previous film-formation process.

In Equation (2), the coefficient A is a
constant pertinent to the apparatus. The coefficient
A has the dimension of pressure and is determined
experimentally.

25   As it is sufficient in the present
invention that the pressure of the mixed gas, which
is subjected to the source concentration measurement,
is determined, the location of the manometer 18A is
not limited to the one shown in FIG.8. Thus, it is
30   also possible to provide the manometer 18A
immediately before or after the FTIR 40 as
represented in FIG.9.

Further, because the FTIR 40 of the present

embodiment can measure the absolute concentration of the source gas by using the FTIR 40, it is not mandatory to conduct the concentration measurement source gas at the downstream side of the node P1.

5 Thus, it is also possible to conduct the pressure measurement at the upstream side of the node P1 as in the case of FIG.10. In the construction of FIG.10, it should be noted that the foregoing pressure measurement can be conducted by the manometer 18

10 provided on the gas line 30, and the use of additional manometer 18A is avoided.

[MODIFICATION]

Similarly, it is possible to modify the
15 MOCVD apparatus 200A of FIG.3 by applying the manometer 18A as represented in an MOCVD apparatus 200E shown in FIG.11. Thereby, it becomes possible to obtain the absolute concentration of the source gas in the source gas line 30. In FIG.11, those parts
20 corresponding to the parts described previously are designated by the same reference numerals and the description thereof will be omitted.

Further, modifications similar to those of FIGS.9 and 10 are possible also in the MOCVD
25 apparatus 200E of FIG.11.

Further, it is possible to obtain the absolute source gas concentration in the source gas line 30 in the MOCVD apparatus 200B of FIG.4 by adding the manometer 18F as represented in an MOCVD
30 apparatus 200F of FIG.12. In FIG.12, those parts corresponding to the parts described previously are designated by the same reference numerals and the description thereof will be omitted.

- 41 -

It·should be noted that the modification
similar to those of FIGS.9 and 10 is possible also in
the MOCVD apparatus 200F of FIG.12.

Further, as shown in an MOCVD apparatus
5   200G of FIG.13, it becomes possible to obtain the
absolute source gas concentration in the source gas
line 30 by adding the manometer 18A to the MOCVD
apparatus 200C of FIG.4. In FIG.13, those parts
corresponding to the parts described previously are
10   designated by the same reference numerals and the
description thereof will be omitted.

In the MOCVD apparatus 200G of FIG.13, too,
similar modifications as in the case of FIGS.9 and 10
are possible.

15

[SIXTH EMBODIMENT]

FIG.14 shows the construction of the FTIR
40 used in various embodiments of the present
invention.

20   Referring to FIG.14, the FTIR 40 includes a
gas passage 401 having optical windows 401A and 401B,
and mirrors 401a - 401c are provided in the gas
passage 401 for reflecting an optical beam incident
through the optical window 401A one after another.
25   The optical beam thus reflected exits through the
optical window 401B, and there is provided a detector
402 for detecting the optical thus exit from the
optical window 401B.

Further, there is provided an
30   interferometer 403 including therein a fixed mirror
403a, a movable mirror 403b and a semitransparent
mirror 403c outside the optical window 401A, wherein
the interferometer 403 introduces the optical beam

− 42 −

from an infrared source 404 into the gas passage
through he optical window 401A.

  The detector 402 supplies an output signal
to an A/D converter 402A for conversion to a digital
5 signal, and the digital signal thus converted
undergoes high-speed Fourier transform in a computer
402B. Thereby, the spectrum of the gas passing
through the gas passage 401 is calculated as shown in
FIG.7.

10   In the FTIR 40 of FIG.14, it should be
noted that baseline length of the interferometer 403
is changed by moving the foregoing movable mirror
403b while simultaneously detecting the intensity of
the incoming infrared optical beam at the detector
15 402. By applying the high-speed Fourier transform to
the interference pattern thus acquired in the
computer 402B,the infrared spectrum of the source gas
is calculated.

  In the present embodiment, it should be
20 noted that the mirrors 401a and 401c are held on a
base body 401C and the mirror 401b is held on a base
body 401D. Thereby, a temperature sensor 401CT such
as a thermocouple and heaters 401CB, 401CD are
provided in the base body 401C. Similarly, a
25 temperature sensor 401DT such as a thermocouple and a
heater 401DB are provided in the base body 401D. In
addition, while the illustration is omitted, the
optical windows 401A and 401B are also provided with
a temperature sensor and a heater.

30   Because the mirrors that make direct
contact with the gas flow are maintained at a
predetermined temperature in the present embodiment,
the problem of formation of precipitates, which is

caused when the source gas undergoes cooling upon passing through the FTIR 40, is positively avoided. It should be noted that the source gas of $W(CO)_6$ has to be maintained at a high temperature during the

5   transportation for avoiding formation of such precipitates.

In each of the foregoing various embodiments, it is also possible to use a non-dispersion infrared spectrometer (NDIR) 50 shown in

10 FIG.15 in place of the FTIR 40. Thereby, it becomes possible to obtain an output signal within the duration of 1 second. In FIGS.15, those parts described previously are designated by the same reference numerals and the description thereof will

15 be omitted.

It should be noted that the NDIR 50 has a construction similar to that of the FTIR 40 except that the interferometer 403 and the computer 402B for carrying out the high-speed Fourier transform are

20 omitted. Further, a chopper 404A is provided in the optical path of the infrared beam emitted from the optical source 404 for interrupting the infrared beam intermittently. It should be noted that the chopper 404A may be provided at any location of the optical

25 path of the infrared beam traveling from the optical source 404 to the detector 402.

In the NDIR 50 of FIG.15, too, the mirrors 401a - 401c contacting with the gas flow directly are maintained at the predetermined temperature, and the

30 problem of formation of the precipitates is avoided. The NDIR 50 of FIG.15 may be used in place of the FTIR 40 in the construction of FIG.12 as represented in FIG.16. It should be noted that similar

- 44 -

modification is possible also in other embodiments.

Further, the present invention is not limited to the embodiments described heretofore, but various variations and medications may be made
5   without departing from the scope of the invention.

For example, it is possible to provide a turbo molecular pump in the pre-flow line 33 in correspondence to the film-formation process that uses a low vapor pressure source material. Further,
10 the diameter of the pre-flow line 33 may be optimized. According to such a construction, it becomes possible to measure the source gas concentration under the condition close to the case of the source gas is actually transported along the source gas line 30 for
15 the film-formation process by using the FTIR 40 provided in the pre-flow line 33.

Further, it is also possible to conduct the detection of the source gas concentration by other means than the FTIR measurement or the measurement of
20 the infrared spectrum. In the case the processing is conducted under a sufficiently high process pressure, it is possible to use the acoustic emission method in view of the relatively high pressure of the source gas. In this case, too, it is possible to calculate
25 the absolute source gas concentration by applying a pressure correction according to Equation (2).

The present invention is based on Japanese priority application 2002-201532 and 2003-191044 filed respectively on July 10, 2002 and July 3, 2003,
30 the entire contents thereof being incorporated herein as reference.